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HIGH-TEMPERATURE METHANE FUEL CELLS

By

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INTRODUCTION

If it were feasible to utilize methane in fuel cells, the development of small power packs for dwellings would be an attractive venture. In many areas of the United States, the cost to the residential consumer of a unit of energy in the form of natural gas (methane) is only one-sixth to one-eighth as much as a unit of energy in the form of electricity. (2, 15) This basic cost advantage would seem to be large enough to offset the unavoidably higher costs of generating electricity in a multiplicity of small installations.

Direct electrochemical oxidation of methane does not appear practical at this time, because of low reactivity in all types of fuel cells. However, it is possible to use it "directly" in a high-temperature fuel cell in the presence of steam. Reforming is presumed to occur on the anode surface, with the observed current due primarily to electrochemical oxidation of intermediate hydrogen. (8, 9)

As an alternative, a hydrogen-rich stream can be produced from methane in external apparatus by such gas conversion processes as catalytic steam reforming or partial oxidation, supplemented by carbon monoxide shift if necessary. If caustic electrolytes were to be employed, the effluent could then be fed to a hydrox fuel cell after carbon oxide removal. The removal step would be unnecessary if hydrox cells were selected that carry the charge through the electrolyte via hydrogen ions. In principle, cation exchange cells could be employed, although insensitivity of the catalysts to carbon oxides would have to be established. Recent data on phosphoric acid cells indicate that they may also be considered, although they have received relatively little attention thus far. (14)

Because catalytic steam reforming and partial oxidation of natural gas are widely practiced, and cation exchange cells have been brought to an advanced stage of development, a power pack based on indirect utilization of methane should be technically feasible. However, it seems likely that this approach may be uneconomic because of high investment costs of ion exchange cells (1) and gas conversion and purification equipment.

The technical feasibility of utilizing methane and steam in high-temperature cells has not been thoroughly established in previous studies which were directed primarily to the use of hydrogen and town-gas. (8-11) Considerable improvement in performance of high-temperature methane cells should be achievable. This would not be as likely for the gas conversion processes and low-temperature cells of the indirect approach, which have received far more attention.

PREVIOUS STUDIES OF HIGH-TEMPERATURE FUEL CELLS

Almost all of the recent work on high-temperature cells has involved molten carbonate electrolytes. Hydrogen, carbon monoxide and propane have been the principal fuels, and a few tests with methane have been reported. (8-11) The technology includes: 1) molten electrolyte held in porous sintered magnesia matrices, (8, 9, 11, 16, 17, 23) 2) plastic ("pasty") mixtures of molten electrolyte and inert filler (10, 11), and 3) free molten electrolyte. (11-13) In practical cells, powder electrodes and single-porosity sintered electrodes have been used with the first two electrolyte-retention methods, (8-11, 16, 17, 23) and dual-porosity sintered electrodes with the last. (11, 12)

Nickel has been the principal anode material. (8-10, 12, 13, 16, 17, 23) Silver (8-10 (8-10, 12, 13), silverized ZnO (11), and lithiated NiO (16, 17, 23) were shown to be

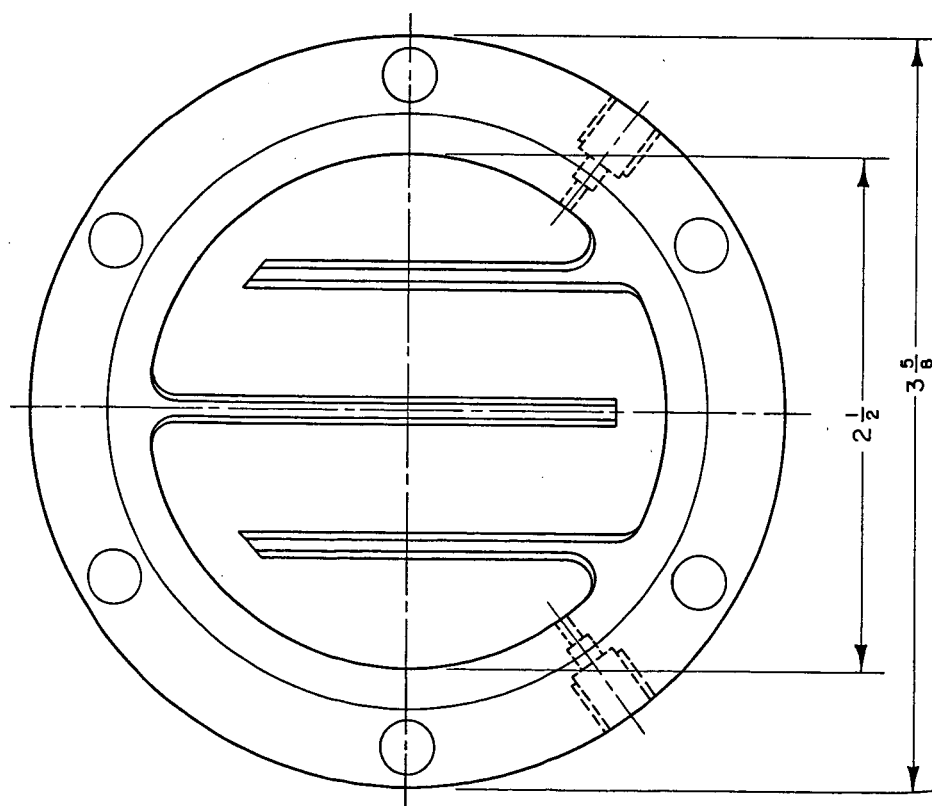
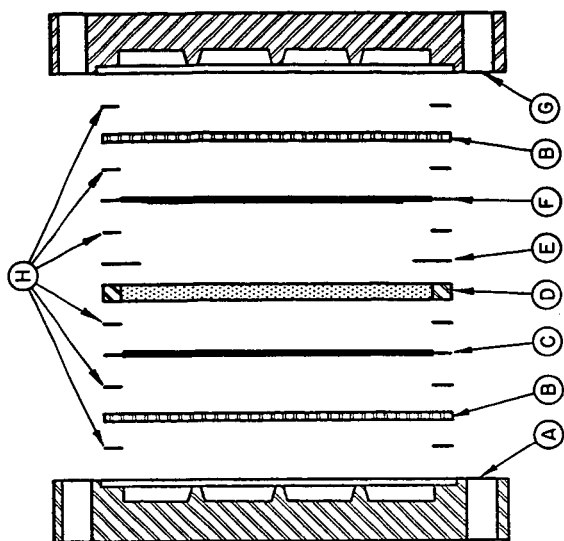


Figure 1. Fuel cell flanges were constructed with fingers to provide contacting pressure and distribute gas over electrode surfaces.



A ANODE FLANGE

B PERFORATED PLATE, 2-3/4" OD x 1/32" THICK

C POROUS NICKEL ANODE (CLEVITE),

COINED ON BOTH SIDES, 2-3/4" OD x 50 MIL THICK

D STEEL RETAINING RING FOR PLASTIC

ELECTROLYTE, 2-3/4" OD x 2-1/2" ID x 1/8" THICK

E MICA SEPARATOR, 2-3/4" OD x 2-1/4" ID

F POROUS SILVER CATHODE (CLEVITE),

COINED ON BOTH SIDES, 2-3/4" OD x 50 MIL THICK

G CATHODE FLANGE

H SILVER GASKET, 2-3/4" OD x 2-1/2" ID x 0.01" THICK

Figure 2. These components were used in high-temperature fuel cell tests with molten carbonate electrolytes

effective cathode materials. With hydrogen-oxygen cells, little or no polarization has been reported for both anodes and cathodes at operating temperatures above the range 500-600°C. (9, 13).

High-temperature fuel cell studies have also been conducted with solid oxides as electrolytes, by Baur. (3, 4) The most interesting of numerous oxide mixtures tested empirically were "Nernst-mass" (85 mole % ZrO_2 -15 mole % Y_2O_3), and a mixture of 1 part CeO_2 , 3 parts WO_3 and 2 parts clay. Baur's efforts were not successful from a practical viewpoint. Carbon was the fuel, which is much less reactive than conventional fluid fuels. Electrolytes were formed into thick-walled thimbles, and fuel cells were constructed in the form of crucibles. No attempt was made to build cells with practical shapes or thicknesses. Further, oxides of polyvalent elements such as cerium were studied, which may exhibit semiconductivity at fuel cell operating temperatures, and cause cell short-circuiting. In recent years, some interest in solid oxide electrolytes has been noted (5), but the concepts have not yet been demonstrated with gaseous fuels in practical cells, as has been done in the case of molten carbonate electrolytes.

In the present work, the effects of methane flow rate were studied in leak-tight cells at 650° and 750°C. Comparisons of molten carbonate cells were made, using: 1) plastic mixtures of carbonates and powdered magnesia, and 2) porous sintered magnesia matrices impregnated with molten carbonates. Some preliminary results of a study of the effects of temperature from 750° to 850°C, have been published. (25)

CARBONATE ELECTROLYTES

Experimental

Disk-shaped fuel cells were constructed with pressure fingers, to give good contacting and gas distribution (Fig. 1). The components for plastic electrolyte tests and the silver gasket sealing technique are shown in Fig. 2. Details of the construction have been published. (25)

Porous sintered nickel and silver electrodes were fabricated with embedded metal gauzes for strength. The adaptation of nickel-cadmium battery plaque techniques to high-temperature fuel cell operation was carried out by the Clevite Corporation.

Plastic electrolytes were prepared from -325 mesh, high-purity (99.0%) crystalline magnesium oxide and the equimolar eutectic Na LiCO₃. Mixtures of the powders were cold-pressed in steel retaining rings shown in Fig. 2, and heated to just above the melting point of the eutectic. After cooling, the disks were ground flush with the retaining ring. Electrolyte flaws were common, and could be readily patched by applying additional mixed powders and reheating.

Cell short-circuiting through the metal retaining ring was prevented by use of a thin ring of mica which performed satisfactorily in short tests. Over extended periods this would not be feasible due to loss of water from the mica, so experiments with steel retaining rings flame-sprayed with insulating ceramics have been initiated with good results.

Magnesia matrices were prepared initially by a method used by previous investigators. (23) Improvements which essentially eliminated warpage were made with the aid of the Ceramics Research Division of Armour Research Foundation. Disks were impregnated with molten NaLiCO₃, cooled, and ground to measure 0.118 ± 0.003 inches thick at a number of points. The cell assembly and sealing technique was identical to that shown in Fig. 2, except that the impregnated disk replaced the retaining ring, plastic electrolyte and mica.

The sealing method was successful in eliminating peripheral leakage of gases. The plastic electrolyte technique eliminated cross-leakage of oxidant and fuel also, as demonstrated by inclusion of helium and argon tracers in the feed streams, and mass-spectrometric analysis of the exit gas streams. In cells with ceramic matrices, it was not possible to prevent cross-leakage completely.

Fuel and oxidant were fed as mixed gases supplied by the Matheson Company in cylinders. A small amount of carbon dioxide was included in the fuel to insure against electrolyte decomposition, and the stoichiometric CO_2/O_2 feed ratio for carbonate ion formation was used in the oxidant stream. Typical analyses of the gases were:

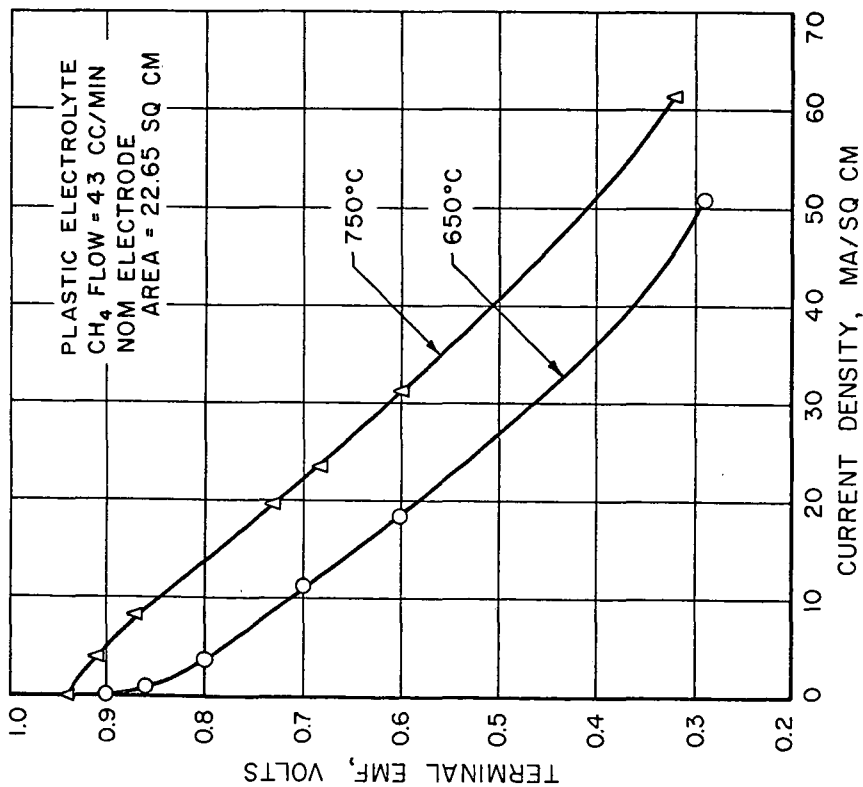


Figure 3. With steam-methane feed gas, better results were obtained at 750° than at 650° C, but performance level was still of practical interest at 650° C

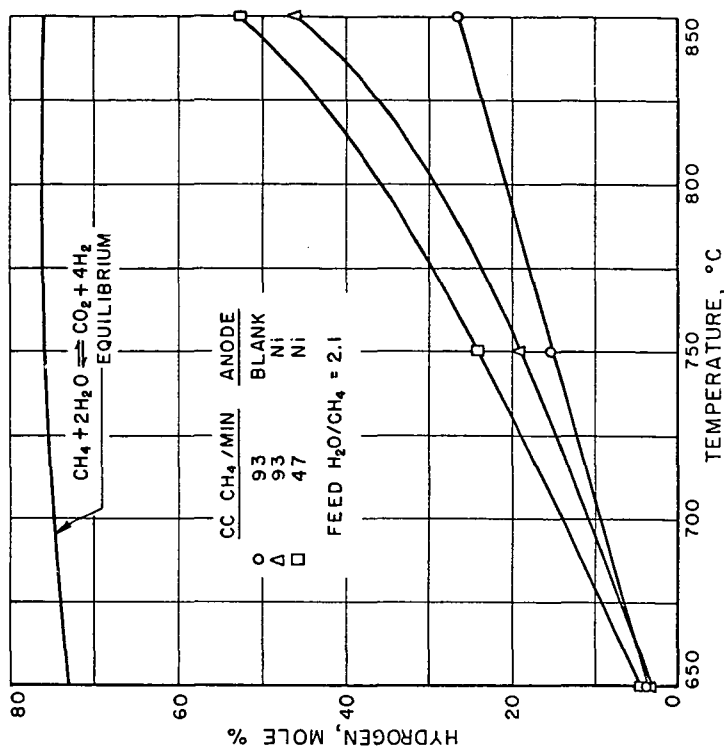


Figure 5. Porous sintered nickel anodes, prepared by commercial methods for nickel-cadmium battery service, were shown to have catalytic steam reforming activity at methane fuel cell operating temperatures

<u>Mole %</u>	<u>Fuel Stream</u>	<u>Oxidant Stream</u>
CH ₄	92.0	--
C ₂ H ₆	0.3	--
Higher Hydrocarbons	0.6	--
CO ₂	4.7	65.1
H ₂	0.3	--
O ₂	--	33.3
N ₂	0.5	--
A	--	1.6
He	1.6	--
Total	100.0	100.0

Steam was generated in an electrically heated tee filled with wire packing, and fed through a hypodermic tube. The fuel stream was passed through the heated tee to pick up the steam, before being conducted to the fuel cell. A gas-fired furnace was used to heat the fuel cells.

The flow of water and all gas streams was measured by calibrated rotameters. Water was condensed from the anode exit gas, and the stream was dried by passage over Drierite before flow measurement. Reported gas volumes are in cubic centimeters, dry, at 0°C, and one atmosphere.

Temperatures were sensed by a chromel-alumel thermocouple embedded in one flange of the cell. All tests were conducted at atmospheric pressure.

Cell polarization data were obtained upon stabilization of performance after application of load. A constant-load test showed no perceptible drift in performance with time for 10 to 20 hours. During this period, terminal voltage and current could be measured with good reproducibility.

Feed and exit gas analyses were carried out with a Consolidated Engineering Company Model 21-103 mass spectrometer. X-ray tests of electrode and electrolyte materials were performed on selected specimens with a General Electric X-ray diffractometer.

In all methane tests, the cell was brought to temperature with hydrogen flowing over the anode, and a one-hour hydrogen treatment at 650°C. was given, before the methane-steam mixture was introduced. Also, during the approach to operating temperature, the oxidant mixture was passed continuously over the cathode.

Effects of Variables

Figure 3 shows typical cell polarization curves for 33 mole % methane - 67 mole % steam at 650° and 750°C., using plastic electrolyte. Performance at 650°C. was at a higher level than expected from published observations on the lack of reactivity of methane below about 700°C. (10)

With electrolyte in magnesia matrices, no effect of methane flow rate was observed from 23 to 93 cc/min (Fig. 4). A decrease to 9.3 cc/min resulted in a decline in performance above about 15 ma/sq cm; at this current density, the flow rate corresponds to about eleven times the stoichiometric requirement. At the highest current density obtained at 9.3 cc/min (44.2 ma/sq cm), the flow rate is about four times the stoichiometric.

Figure 4 also compares the performance electrolyte mixture, with the more conventional magnesia matrix. A substantial improvement was obtained with the plastic electrolyte, amounting to about 40% more current at 0.5 volts, and higher percentage increases at higher voltages.

A significant amount of electrolyte penetrated the electrodes during gaseous cell tests. For example, carbonate analysis of a typical used magnesia matrix indicated that about 24% of the initial charge of electrolyte had been lost. To see if this was due to seepage into the electrodes, or due to decomposition of the carbonates, the matrix was also analyzed for sodium and lithium. The observed carbonate content agreed well with the value based on the assumption that all sodium and lithium was present as carbonate. Further, analysis of the electrodes indicated presence of roughly the amount of carbonate lost from the matrix.

In one test, the consequences of loss of methane flow were explored. Steam flow was maintained, and methane was replaced by carbon dioxide; performance dropped almost immediately. After 1-1/2 hours, short-circuit values of only 0.06

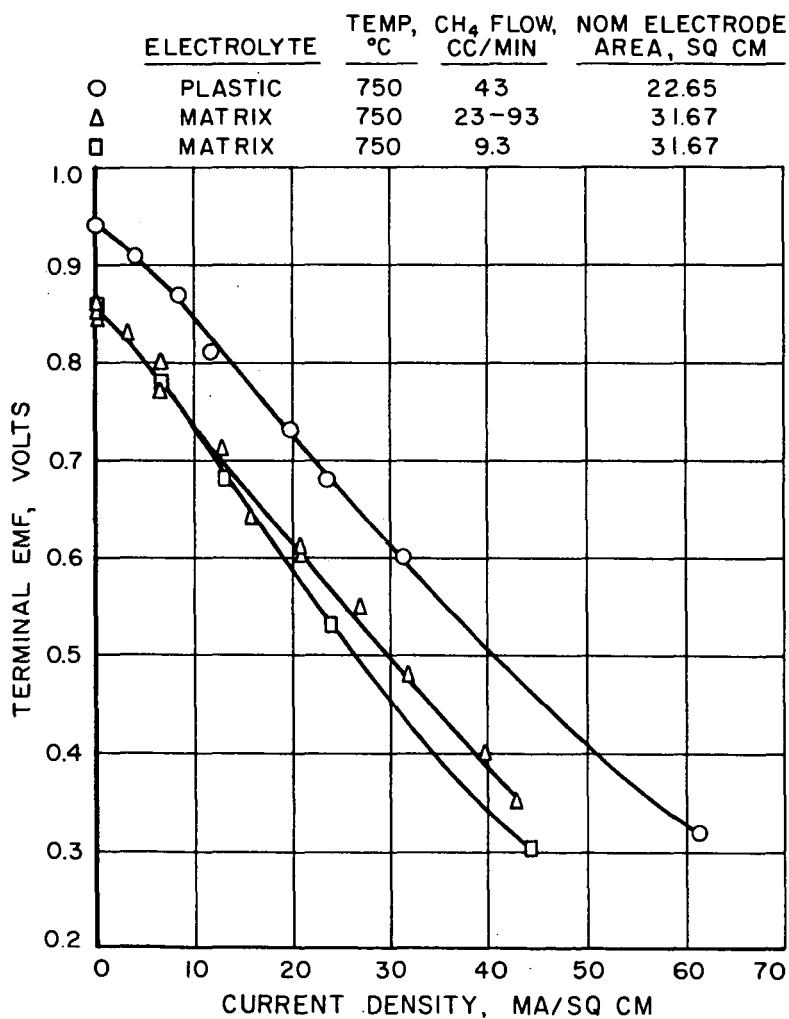


Figure 4. Plastic electrolyte mixtures of magnesia and molten carbonate showed superior performance, as compared to carbonate held in sintered matrices. Little effect of flow rate was noted below about 15 ma/sq cm current density

volts and 5.7 ma/sq cm were obtained. Hydrogen was then fed for two hours, but performance was not restored. Examination of the cell after the run indicated that most of the electrolyte had been lost from the matrix, and that the perforated disk supporting the anode had been partially fused in place by heat. X-ray studies of the anode showed strong lines for nickel oxide, as well as NaLiCO_3 and nickel; normally no lines for nickel oxide are found. Thus, it appears that sudden loss of methane flow can permit rapid oxidation of the anode, localized overheating, and expulsion of electrolyte.

Catalytic Reforming Tests with Anodes

Porous sintered nickel anodes cut from battery plaques manufactured by Clevite Corporation for nickel-cadmium batteries, were tested for steam reforming and CO shift activity at temperatures from 650° to 850°C. Runs were conducted using fuel cell flanges, but with the electrolyte and cathode replaced by a steel plate.

Methane flow rates of 47 and 93 cc/min (1.5 and 2.9 cc/min-sq cm electrode) were studied, with a steam to methane molar feed ratio of 2.15. Flow per unit area may be compared with industrial steam reforming practice for methane; for wide-packed, 1/2-inch spheres of supported nickel catalyst, a value of about 3 cc/min-sq cm catalyst surface gives good conversion at 750° to 850°C.

The results shown in Figure 5 indicate low activity at 650°C., increasing significantly with temperature. The decrease in flow rate from 47 to 93 cc/min led to some increase in conversion, at each temperature studied. In additional tests (not shown) significantly higher catalytic activities were often achieved by prolonged pretreatment with hydrogen at 650°C. before the methane-steam mixture was introduced. Only a one-hour treatment was used in the tests of Figure 5.

In one 750°C. reforming test at 100 cc/min (not shown), no steam was fed, and the methane feed stream was dried by passage through two dry ice-acetone condensers in series. Considerable carbon formation occurred. After five hours, the anode was swollen, very friable, and contained 52.3 wt. % carbon by combustion train analysis. Nickel content was 47.8 wt. % by dimethylglyoxime precipitation.

X-ray examination showed graphite and nickel, but no carbides. Gas samples taken after one and four hours at temperature contained 20.0 and 31.0 mole % hydrogen, respectively, indicating substantial activity for methane cracking without the presence of steam.

OXIDE ELECTROLYTES

Although molten carbonate electrolytes are useful, they may eventually be replaced by electrolytes which will avoid some or all of the inherent disadvantages of carbonates. Molten carbonates require a finite partial pressure of carbon dioxide at both anode and cathode, to prevent decomposition. Also, carbon dioxide must be included in the oxidant feed gas in near-stoichiometric quantities for the generation of carbonate ions. Further, carbonates are corrosive, have low viscosities and are difficult to retain over long operating periods.

Porous sintered magnesia matrices are commonly used to contain molten carbonates, and a substantial thickness is required to give adequate mechanical strength to the matrix. The presence of the matrix leads to ionic conductivities at least an order of magnitude lower than those for the same thickness of free fused salt. (10) It is difficult to impregnate all pores of sintered matrices, and cross-leakage of gases between anode and cathode rooms is usual. (9) Further, in vertical configurations, electrolyte flows within the matrix due to gravity, tending to open pores, increasing cross-leakage and the non-electrochemical reaction of fuel and oxidant. If free electrolyte is used, electrode flooding can occur unless expensive dual-porosity electrodes are employed.

For these reasons, a solid oxide electrolyte was sought to transport oxygen in the form of oxide ions. An approach to the following ideal properties was desired:

1. Oxide ion transference number of unity.
2. Rapid migration of oxide ions, since current density is directly proportional to rate of oxide ion migration.
3. Chemical stability in the presence of reducing gases or oxygen at temperatures as high as about 1000°C.

4. Physical stability, with no shrinkage, creeping or warping in service.
5. Impermeability to gases.

The migration of oxygen ions through many solid oxide electrolytes can be described by Fick's law of diffusion, with the diffusion coefficient represented as an Arrhenius function of temperature. (21) High permeation rates are favored by increases in temperature and concentration gradient. The highest concentration difference that can be realized will be fixed by the concentration of oxygen on the cathode surface, and an oxygen concentration of zero at the anode. If this obtains, then permeation rate can only be increased by increases in temperature or decreases in electrolyte thickness, for a given electrolyte.

Previous attempts to use solid oxide electrolytes in fuel cells (3, 4) resulted in permeabilities too low for practical interest, largely due to excessive electrolyte thickness. At that time, the diffusion mechanism of oxygen ion migration in solid oxides had not yet been clarified.

Theory of Ionic Conduction in Oxides

Oxygen may migrate via interstices, or via oxygen ion vacancies in the crystal lattice. Relative sizes of the atoms or ions, and the type of crystal structure, control the mode of diffusion. In fluorides having the fluorite structure, Seith has noted that the anion is the only species to migrate. (24) The similarity in radius between fluoride and oxide ions suggests oxide ion diffusion only, in fluorite-type oxides. Kingery has demonstrated this for $Zr_{0.85}Ca_{0.15}O_{1.85}$, by means of heavy oxygen exchange experiments, and electrolytic conductivity measurements. (21)

Because of the difference in valence, each calcium atom combines with only one oxygen atom instead of the two held by zirconium. The fluorite structure may be preserved with a given stoichiometry, by creation of oxygen ion vacancies, or addition of interstitial cations; density measurements indicate that the former prevails. Therefore, Kingery concluded an oxygen ion vacancy mechanism accounted for the conduction in $Zr_{0.85}Ca_{0.15}O_{1.85}$, and postulated that conductivity increases proportionately to the concentration of oxygen ion vacancies, at low concentration levels. (21)

A number of materials can generate vacancies in the lattice. This can occur, for instance, when cations of valence less than four replace zirconium randomly in the fluorite lattice. The term "mixing oxide" is used to denote additions of this type, and examples are the oxides of calcium, magnesium, yttrium, scandium, lanthanum and other rare earths. In this instance, zirconia may be termed the "host" oxide; other hosts are the oxides of hafnium, thorium, uranium, and rare earths such as cerium, praseodymium and terbium. However, the tetravalent rare earths and uranium may become semiconductive at high temperatures. Also, there is evidence that thorium systems show electronic conductivity, perhaps due to impurities; under the same conditions, ZrO_2 -CaO solid solutions were not semiconductive. (22)

Conductivity Studies on Solid Oxides

Conductivity measurements for $Zr_{0.85}Ca_{0.15}O_{1.85}$ have been published. (18, 21, 26) Since current is carried only by oxygen ions, oxygen permeation values can be converted to current density values. For example, to obtain 100 ma/sq cm, temperatures from 700° to 1000°C., and electrolyte thicknesses from one to 10 microns are indicated, assuming that all lattice vacancies are filled at the cathode-electrolyte interface, and empty at the anode-electrolyte interface. Limitations in electrode performance, or in electrode-electrolyte contacting would lead to lower performance levels, of course.

Trombe and Foex (26) studied the conductivity of the zirconia-calcia system, as a function of composition at 1000°C. In contradiction to Kingery's postulation, the maximum conductivity (about 250 ohm⁻¹cm⁻¹) was found at about 15 mole % calcia, at the phase boundary between monoclinic zirconia saturated with calcia, and the fluorite-type solid solution with the minimum amount of calcia. The decrease in conductivity in the fluorite region from 15 to 33 mole % calcia was attributed to ordering in the lattice. The conductivity of several solid solutions in the thoria-yttria and thoria-lanthana systems have also been measured, and values of practical interest to fuel cell applications recorded. (19, 20)

Fabrication of Thin Films of Oxides

To obtain current densities of significance, thin films are essential. A number of useful techniques are being evaluated for this purpose: flame spraying (6), electron beam vaporization, and solution ceramics, (7) Preparation of thin wafers by conventional pressing, and optical grinding and polishing methods, does not appear adequate.

The application of sufficiently thin (about one micron) and even coatings of $Zr_{0.85}Ca_{0.15}O_{1.85}$ on porous sintered metal electrodes was carried out for the Institute by the Alloyd Corporation. The technique involves impingement of a beam of high-energy electrons on a target of oxide, vaporization of the oxide, and condensation of the vapors on a cool substrate. In these tests, the films were porous and not sufficiently adherent. Some changes in composition during vaporization were found by X-ray examination, but the fluorite structure was preserved.

Flame spraying may have more promise, and is presently being investigated for the Institute by the Ceramics Research Division of Armour Research Foundation. In this approach, fine particles of oxide are entrained in the oxygen stream of an oxy-acetylene or oxy-hydrogen flame, and molten or semi-molten particles are directed against a substrate. Porosity may be overcome by applying suitable impregnants, or by densifying the flame-sprayed wafer. Porous layers of metal electrode materials may then be applied to either or both sides of the wafer, also by flame-spraying, to give intimate electrode-electrolyte contact and excellent adherence. In a variation of this method, the oxide powder is fed into the gas used to stabilize the arc of a plasma jet.

Preliminary fuel cell tests of wafers of $Zr_{0.85}Ca_{0.15}O_{1.85}$ prepared by flame-spraying, with and without flame-sprayed electrodes, have given encouraging results. By flame-spraying, electrolyte films cannot yet be prepared as thin as by electron beam vaporization. Indications are that it may be possible to reduce thickness sufficiently to obtain practical current densities, in accordance with the predictions based on published conductivity measurements.

UTILIZATION CONCEPTS

A natural gas power pack for home installation would provide for the addition of steam to the gas feed, and in the case of carbonate electrolytes, the addition of flue gas as a source of carbon dioxide to the air feed. Waste gas from the cells would be under-fired to compensate for normal heat losses. Waste heat could be utilized in several conventional appliance functions in the home.

The high operating temperature of methane fuel cells is not compatible with the on-off, low load factor character of home utilization of electricity. Storage batteries could be used to raise the load factor so the high-temperature fuel cells could operate essentially continuously. Thus, investment in a large pack of fuel cells capable of meeting peak electrical loads would be unnecessary.

CONCLUSIONS

With methane-steam feed at low flow rates, it is possible to obtain current densities of practical interest, using plastic electrolyte mixtures of $NaLiCO_3$ and powdered magnesia in flat cells having porous sintered nickel anodes and porous sintered silver cathodes. Electrode fabrication techniques of commercial nickel-cadmium battery practice are adaptable to high-temperature fuel cell service, and steam-reforming tests of commercial sintered nickel anodes indicate considerable catalytic activity for methane conversion to hydrogen.

Substantially higher performance levels can be achieved with plastic electrolyte, than with electrolyte held in porous ceramic matrices. Further, cross-leakage of gases between anode and cathode rooms can be eliminated with plastic electrolyte, but is difficult to eliminate with the ceramic matrix technique. Peripheral leakage in flat-disk cells can be avoided by use of coined electrodes and silver gaskets, and steel retaining rings for plastic electrolytes.

In principle, development of solid oxide cells would be desirable, because of the inherent limitations of molten carbonates. It is proposed that a number of solid state reaction products with the fluorite structure should be useful as replacements for carbonates, in high-temperature fuel cells. Thin films are necessary, and promising results have been obtained with flame-sprayed films of oxides, with and without flame-sprayed nickel electrodes.

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